

Interactive comment on “SCIAMACHY formaldehyde observations: constraint for isoprene emissions over Europe?” by G. Dufour et al.

G. Dufour et al.

Received and published: 6 February 2009

The authors thank the referees for their interest in the article and their suggestions for improvements. The comments made are addressed below.

One major point has been underlined by the both reviews: the current manuscript does not look complete and does not answer the question posed by the title without including explicit inverse modelling results. The authors agree that the manuscript can appear unfinished especially in regards of the title. Since the submission of the present paper, additional work has been done in order to finalize a first inverse modelling experiment with the spatial and temporal averages prescribed here. Rather than changing the title of the current paper and present inverse modelling results in another paper, the authors

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have decided to follow the recommendation of the referees and to add preliminary inverse modelling results to make the current paper more valuable. The original section 5 has then been replaced by a section presenting first inverse modelling results. This section consists of a brief presentation of the applied method, of the presentation of the first results obtained and a brief discussion of these results.

In the following, the authors will answer to the specific comments of each referee separately.

REPLY TO REFEREE #1:

The referee expressed his regrets in the review concerning the lack of accuracy in the wording and the lack of detail in some parts of the text. The authors have tried to improve these points as much as possible, especially for the parts underlined by the referee.

REPLY TO COMMENT #1:

"The title is inaccurate: if only we could constrain emissions using space-borne data. More accurately, data constrains emission estimates".

The authors agree that the actual title could be ambiguous and they have changed it as suggested by the referee. The new title is:

"SCIAMACHY formaldehyde observations: constraint for isoprene emission estimates over Europe ?".

REPLY TO COMMENT #2:

"Abstract, line 7. Europe has been studied before! The authors mean that satellite observations of formaldehyde over Europe have not previously been studied".

The phrase "never studied before" has been removed at line 7 of the abstract. The sentence "Europe was never specifically studied before for these purposes using satellite observations" has been added at line 10 after "HCHO columns".

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REPLY TO COMMENT #3:

"Page 19276, line 26. According to the ESA website, SCIAMACHY should be spelt out as the Scanning Imaging Absorption SpectroMeter for Atmospheric CHartography".

The spelling of the SCIAMACHY acronym has been checked and corrected.

REPLY TO COMMENT #4:

"Section 3: the tagging scheme is a substantial piece of work. Has this work been published previously? If no, the authors are obliged to describe it in a bit more detail. They seem to make some simplifications to the Pfister scheme that needs to be clarified"

The work on the tagging schemes implemented in the model has not been published before. The authors now provide more details about the method used. In particular, they added a table that summarizes the tagging technique with respect to the reaction type. They also provide more details on how they have treated the 4th pathway corresponding to the tagging of anthropogenic species. They added the following part at line 23 of page 19283:

"In the case of the 4th pathway, several HCHO precursors with anthropogenic sources included within the CHIMERE chemical mechanism are treated simultaneously. Only those directly emitted without any secondary sources are considered, in order to avoid too strong complication of the tagged scheme. In particular, CH₃CHO, CH₃OH and MEK are emitted but can also be produced secondarily, and thus are not considered"

REPLY TO COMMENT #5:

"Section 3: ignoring the CH₃OH source is fine if the authors can show that this does not compromise their interpretation of the formaldehyde columns. In the current draft they suggest (not based on calculation) that ignoring CH₃OH will introduce a 10% error in reproducing the formaldehyde columns. They should use the model to prove this is indeed the case".

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The use of the model to prove that ignoring biogenic emissions of methanol introduces an error of 10% on the formaldehyde columns would be difficult as the model version used is not set up to include methanol biogenic emissions. However, the assessment of the 10% error introduced when ignoring CH₃OH was based on a separate calculation. The authors use the work done with the box model to evaluate the potential production of HCHO from various precursors including biogenic methanol, taking into account their average emission over the European domain. The 10% errors have been deduced from this calculation. This has now been stated more precisely in the text as follows:

"To estimate the impact of neglecting biogenic methanol on the HCHO column, we calculate the total potential productions of HCHO as the sum of the individual potential production of each precursor (defined as the yield of the precursor multiplied by the corresponding mean European emissions). The total potential production of HCHO obtained when biogenic emissions of methanol are accounted for is about 10% larger than when they are neglected."

REPLY TO COMMENT #6:

"Section 3: in general, correlating emissions and tagged formaldehyde columns does not prove or disprove a limited smearing effect unless the authors have tagged contributions from particular emitting grid boxes"

The sentence "This reflects that the smearing effect is relatively limited" has been reworded to be more precise and now reads:

"This reflects the fact that isoprene is oxidized rather quickly to HCHO, i.e. largely within the same grid-cell".

The influence of emissions from one grid boxes on other parts of the European domain is now discussed in section 5 in terms of the influence of one subdomain onto the others deduced for the observation matrix. This matrix has been built with a perturbative method for the inverse modeling. We show that the off-diagonal terms of the matrix are

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small compared to the diagonal terms (maximum one third but often negligible) and then that the influence of one subdomain on another is limited and often negligible.

REPLY TO COMMENT #7:

"Page 19289, line 8: observations over sea might be "questionable" but why? In any case, the authors are not using the BIRA dataset. How does the Bremen formaldehyde product compare with the BIRA product? And what are the associated implications of these differences for inferring emissions over Europe?"

Since independent correlative measurements of HCHO above water bodies are sparse, it is up to now not clear whether the elevated levels seen by the satellite retrievals above some oceanic regions are real or not. In Wittrock (2006, pp. 149), GOME HCHO data from fall 1996 have been compared to in situ measurements carried out onboard the German research vessel Polarstern. Here a reasonable agreement was found. But as pointed out in the same study and in Wittrock et al., 2006 the retrieval quality for minor trace gases above the oceans is reduced due to spectral interference with several effects: vibrational raman scattering, liquid water absorption, different chlorophyll absorptions, fluorescence from colored dissolved organic matter, Therefore it is out of the scope of this study to discuss the results above water in detail. We have changed the text slightly to make this clear to the readers.

Comparisons between different satellite data sets have been carried out by the authors and also from members of the BIRA group. Wittrock (2006) has found a good agreement between his results and those from the TEMIS project for September 1997 (correlation 0.8 and slope 0.97 on a global scale). DeSmedt et al. have reported an agreement within 10% between the Bremen and the BIRA HCHO products for source regions. Significant differences have been reported for desert regions which is not relevant for this study. For summer 2003 above Europe both products yields similar results but with BIRA somewhat noisier due to the fact that there have used only a subset of the SCIAMACHY spectra available.

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REPLY TO COMMENT #8:

"Page 19290, line 4: there is a shocking lack of detail regarding tree species. The authors should not be shy with the details"

In section 2.1 (Model description), an additional sentence was added to better explain the biogenic emission calculation scheme and the use of country averaged tree species following Simpson et al. (1999):

"The emissions potential depends on land-use and a country averaged tree species distribution, distinguishing several tenths of isoprene and/or terpene emitting tree species (Simpson et al., 1999). In the absence of specific data on tree species distributions over North Africa, the distribution from Greece was used. This introduces additional uncertainty in the emission calculation for this region (see below)."

REPLY TO COMMENT #9:

"Page 19290, line 19: I do not understand this statement".

The sentence has been reworded as follows:

"Additional spatial averaging of regions with similar formaldehyde columns and type of emissions helps to reduce the measurement noise and to have more pertinent comparisons with simulations".

REPLY TO REFEREE #2:

REPLY TO COMMENT #1:

"Page 19274, Line 7: Change to something like "on Europe, a region studied very little so far "".

See the reply to comment #2 of Referee 1.

REPLY TO COMMENT #2:

"Page 19279, Line 2-3: TUV in your moel does not consider the effects of clouds and
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aerosols?"

Clear sky photolysis rates are tabulated from TUV calculations? They are explicitly corrected for cloudiness obtained from ECMWF, but not for aerosols. This is précised in the following text:

"The photolysis rates are calculated using the troposphere ultraviolet and visible model (TUV) (Mandrovich and Flocke, 1998) and and are tabulated depending on altitude and zenith angle. They are corrected for cloudiness, using cloud cover data for low, medium, high and convective clouds delivered by ECMWF."

REPLY TO COMMENT #3:

"Page 19282, first two paragraphs: You describe the different contributions to formaldehyde, but it would be good to also visualise them. I suggest adding in Figure 3 also the model contributions from background, and isoprene and terpene oxidation".

Figure 3 has been modified to include the different contributions to HCHO at each station.

REPLY TO COMMENT #4:

"Page 19285, Line 22ff: Please specify the correlation coefficients".

The spatial correlation coefficients have been given between the emissions and the tagged columns at page 19284, line 22, line 26 and page 19285, line 1. Temporal correlation coefficients between mean European HCHO columns and either mean tagged columns or mean emissions will be misleading because the temporal variability will be strongly smoothed during the average by insensitive boxes.

REPLY TO COMMENT #5:

"Page 19289, Line 4-7: Sentence needs to be re-written".

The sentence has been re-written as follows:

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"However, some observations have to be taken with caution especially when monthly averages are considered. Actually, their uncertainty can be in the range of their absolute value, for instance in some regions in Spain during June and July for example".

REPLY TO COMMENT #6:

"Table 3: Add the standard deviations to your mean values".

The standard deviations of the mean values have been added in Table 3.

REPLY TO COMMENT #7:

"Table 4: Why do you specify the standard deviation for the observations and not also for the model values?"

The standard deviations for the observations were given to show the significance (or lack of it) of the observations. The authors thought that the standard deviations for the simulation were not informative for this purpose. However they do not have any objection to include them in the Table 4 where they now have been added.

REPLY TO COMMENT #8:

"Figure 4: The caption does not seem to belong to this figure".

The authors apologize for this. A mistake occurred during the proof reading procedure. The caption of Table 4 had also been used as the caption of Figure 4. This will be corrected in the next version of the paper. The right caption of Figure 4 is:

"Mean emissions (in molecule/cm²/s) of isoprene, α -pinene, and anthropogenic VOC considered within CHIMERE for summer 2003.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 19273, 2008.

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